NNSA Laboratory Directed Research & Development

# Innovation Advancing Nuclear Security

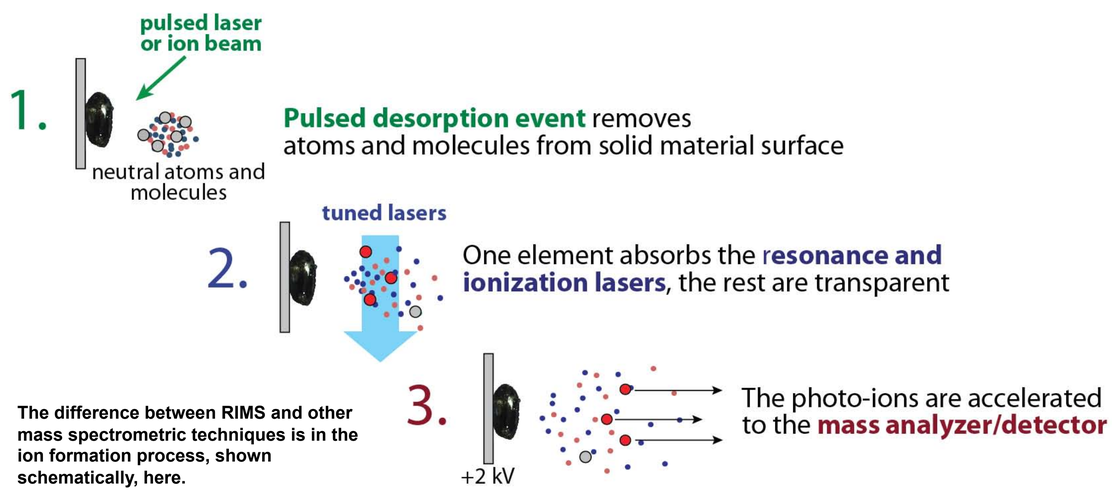
# Resonance Ionization Mass Spectrometry (RIMS) for Nuclear Forensics Applications with Rapid Response

**Kim B. Knight *with* Ian D. Hutcheon (LLNL), Michael R. Savina (ANL), Stan G. Prussin (UCB)**

## RIMS performs chemical separations inside the instrument

- **Post-detonation nuclear forensics demands rapid answers**
- **Sample dissolution and chemical separations necessary for isotopic analyses are the most time-consuming steps in conventional mass spectrometry**
- **RIMS can circumvent lengthy preparation steps, enabling rapid data acquisition**

**In partnership with Argonne National Laboratory and the University of California, Berkeley, we are exploiting the unique ability of Resonance Ionization Mass Spectrometry (RIMS) to image and analyze raw materials *directly and without isobaric interference* to develop a new tool for nuclear forensics. Faster analytical times are the goal of RIMS.**



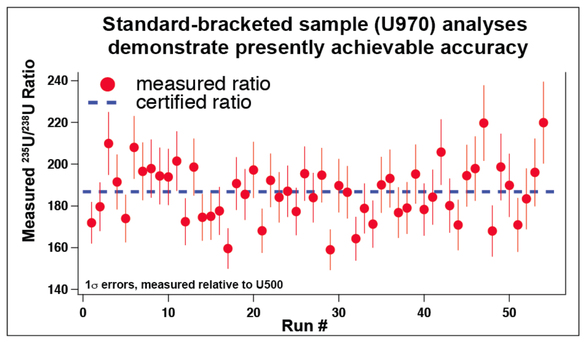

**The difference between RIMS and other mass spectrometric techniques is in the ion formation process, shown schematically, here.**

**Isotope ratio mass spectrometry of the actinide elements is an important technique for nuclear forensics and attribution. RIMS provides isotopic analysis of debris and environmental materials associated with a nuclear detonation without sample purification, using laser spectroscopy to discriminate against isobars (*i.e.* same-mass interferences) in the mass spectrometer.**

## RIMS is a breakthrough in rapid isotope analysis for nuclear forensics

- **Precision and accuracy in $^{235}U/^{238}U$ of better than 1% have been established, applying conventional standard–sample bracketing methods**
- **Excellent selectivity between U and Pu has been confirmed**
- **U and Pu isotopes have been successfully measured in fallout debris samples**

**Measurements of samples enriched in $^{235}U$ from 1–98% demonstrate sub-percent precision over a dynamic range of three orders of magnitude, with no significant deviations from certified standard values.**

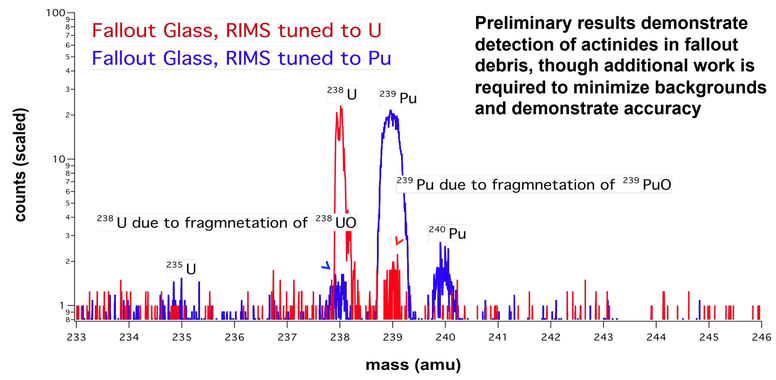



| CRM970 ($U_3O_8$) | |
|---|---|
| $^{235}U/^{238}U_{cert}$ | = 186.77 |
| $^{235}U/^{238}U_{meas}$ | = 185.16 |
| (1σ) | ± 1.76 |
| $\chi^2$ | = 1.10 |

***Standard-sample bracketing now permits sub-percent accuracies on $^{235}U/^{238}U$ ratio measurements over time-scales of hours***

**Analysis times to collect $^{235}U/^{238}U$ and $^{234}U/^{238}U$ data are less than 4 hours per sample, including sample preparation. This work has been extended to multi-actinide samples (mixed U and Pu).**

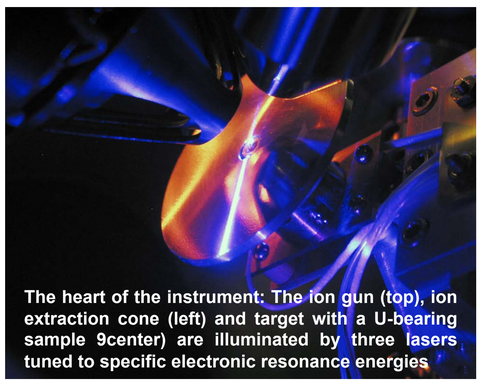
**Work to analyze glassy materials with low (tens of parts per million) actinide concentrations, demonstrates the utility of RIMS to materials such as fallout debris.**




**Preliminary results demonstrate detection of actinides in fallout debris, though additional work is required to minimize backgrounds and demonstrate accuracy**

## Sensitive, selective detection of uranium: Laser control is the key

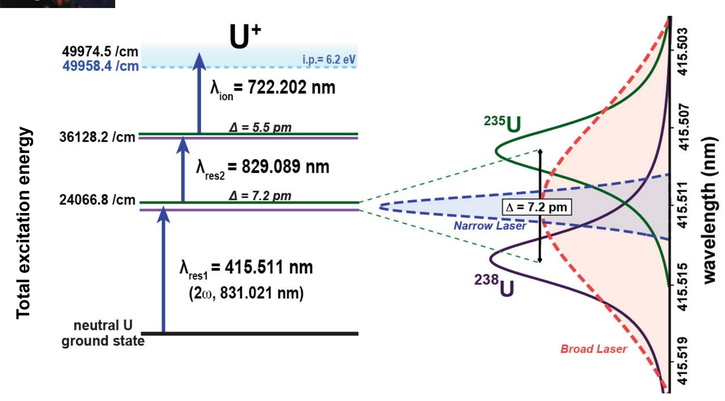
- **Using sample-preparation methods similar to those already in common use, RIMS is capable of ultra-trace analysis as or more sensitive than current techniques on samples containing very few atoms of the element of interest**



**The heart of the instrument: The ion gun (top), ion extraction cone (left) and target with a U-bearing sample 9center) are illuminated by three lasers tuned to specific electronic resonance energies**

**Large isotope shifts, characteristic of deformed nuclei, including actinides, cause very small shifts in wavelength to create large isotopic fractionation, and form a primary challenge to RIMS analysis of actinides.**

**A broadband laser approach (from 1.5 pm to more than 5 pm) reduced the sensitivity of the system to wavelength jitter by an order of magnitude, increasing analytical precision of the $^{235}U/^{238}U$ ratio from ±10% to better than 0.5%.**




**A 3-step resonant ionization scheme for U maximizes uranium sensitivity and selectivity, and shows the effect of laser band width on measured isotope ratios.**

## Actinide isotope analyses by RIMS provides rapid, first-order answers to crucial post-detonation questions

- **Developing RIMS for routine, accurate isotope analysis of actinides provides a powerful new tool for nuclear forensics and attribution**

**This project brings together world-class nuclear forensic capabilities at Lawrence Livermore National Laboratory and years of expertise on RIMS isotopic analysis at Argonne National Laboratory to develop RIMS methods for rapid, high-precision, accurate U and Pu isotopic analysis of post-detonation material using minimal sample preparation.**

**This work is the first-ever systematic study of precision and accuracy in RIMS isotope measurements. A chart of the elements, however, shows that RIMS can be applied to almost any element in the periodic table. Ionization schemes have been demonstrated for most elements (blue), and for many, applied measurements have been reported (green), as well.**

**A RIMS Periodic Table**

accessible by RIMS
published RIMS studies
published RIMS isotopic measurements

| | | | | | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| H | | | | | | | | | | | | | | | | | He |
| Li | Be | | | | | | | | | | | B | C | N | O | F | Ne |
| Na | Mg | | | | | | | | | | | Al | Si | P | S | Cl | Ar |
| K | Ca | Sc | Ti | V | Cr | Mn | Fe | Co | Ni | Cu | Zn | Ga | Ge | As | Se | Br | Kr |
| Rb | Sr | Y | Zr | Nb | Mo | Tc | Ru | Rh | Pd | Ag | Cd | In | Sn | Sb | Te | I | Xe |
| Cs | Ba | * | Hf | Ta | W | Re | Os | Ir | Pt | Au | Hg | Tl | Pb | Bi | Po | At | Rn |
| Fr | Ra | ** | | | | | | | | | | | | | | | |

| | | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| * | La | Ce | Pr | Nd | Pm | Sm | Eu | Gd | Tb | Dy | Ho | Er | Tm | Yb | Lu |
| ** | Ac | Th | Pa | U | Np | Pu | Am | Cm | | | | | | | |

**We continue to refine RIMS techniques for rapid, accurate isotopic analysis of actinides in post-detonation materials, including work to enhance selectivity and maximize useful yield for actinides, benchmarking of RIMS against conventional mass spectrometry, and developing ultra-trace isotope methodologies for actinides.**

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